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THEORY OF DIFFUSION OF RADON AND THORON THROUGH MEMBRANES UNDER STEADY-STATE AND TIME-DEPENDENT CONDITIONS - PART I (STEADY-STATE CASE)

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THEORY OF DIFFUSION OF RADON AND THORON THROUGH MEMBRANES UNDER STEADY-STATE AND TIME-DEPENDENT CONDITIONS - PART I (STEADY-STATE CASE)

by

J. Bigu*

ABSTRACT

The theory of diffusion of radon and thoron through a membrane separating a well-defined region of space (sensitive volume) from the rest (outside volume) is discussed. Expressions for the differential and integrated activities in the sensitive volume, versus time, are provided for the direct diffusion process and the reverse diffusion process under steady-state conditions, i.e., the radon and thoron concentrations in the outside volume remain constant during the diffusion process. The formulae presented here are relevant for practical application in the fields of personal dosimetry and environmental monitoring.

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THÉORIE DE LA DIFFUSION DU RADON ET DU THORON À TRAVERS LES MEMBRANES EN RÉGIME PERMANENT ET EN FONCTION DU TEMPS
PARTIE 1 - (CAS - DIFFUSION EN RÉGIME PERMANENT)

par

J. Bigu*

RÉSUMÉ

Le rapport porte sur la théorie de la diffusion du radon et du thoron à travers la membrane qui sépare un espace bien défini (volume sensible) du reste (volume extérieur). Il présente des formules qui définissent les activités différentielles et intégrées du volume sensible, en fonction du temps pendant les processus de diffusion directe et inverse en régime permanent, i.e., les concentrations de radon et de thoron dans le volume extérieur demeurent constantes pendant la diffusion. Les formules exposées dans le présent rapport se prêtent à une application pratique dans les domaines de la dosimétrie personnelle et du contrôle environnemental.

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INTRODUCTION

The ability to separate materials from each other is an interesting and useful property of certain organic materials usually referred to as membranes. The separation of gases and/or airborne particulates is of particular interest in the radiation field. Radon, thoron and their decay products are known to permeate through membranes of organic materials in varying degrees.

An interesting feature of using organic membranes for separating radioactive species lies in the fact that product separation is accomplished, not only by the different diffusion rate of the diffusing species, but also because of the different radioactive half-lives of the species. Hence, if the rate of diffusion of a given radioactive product is so slow that the time that it would take for the product to diffuse through the membrane by purely diffusional processes is significantly larger than its half-life, virtually complete removal of the product will be realized on the basis of radioactive decay alone during the diffusion process. However, it should be noted that it is quite possible for the daughter product formed from decay of the parent to reach the 'other side' of the membrane, and hence, diffuse through.

Separation of radioactive species based on the combined effect of diffusion and radioactive decay during the diffusion process constitutes a remarkably useful feature of certain organic membranes. The general properties of organic membranes to separate gases, to separate radon and thoron from their decay products, and to separate radon from thoron, has been investigated by several authors (1-6). Some practical applications have been found for these properties.

This paper describes the theory of diffusion of radon and thoron through a membrane which separates a well-defined, relatively small, physical volume, i.e., sensitive volume, from the rest. The paper is divided into two

parts. Part I deals with the theory of diffusion under steady-state radiation conditions. Part II deals with the theory of diffusion under time-dependent radiation conditions. In both cases, analytical expressions are provided suitable for personal dosimetry and environmental monitoring, and for other practical purposes.

THEORETICAL BACKGROUND (STEADY-STATE CASE)

The following items of interest are dealt with below:

1. Diffusion of radon and/or thoron into a sensitive volume, initially free of radioactive gas. This process will be referred to in the future as the direct diffusion process;
2. Diffusion of radon and/or thoron out of the sensitive volume into a region with negligible radon and/or thoron concentration, i.e., radioactive gas-free. This process will be referred to in the future as the reverse diffusion process; and
3. Calculation of radon daughter concentration and/or thoron daughter concentration formed in the direct and reverse diffusion processes.

In order to simplify items 1 to 3, the following assumptions have been made:

1. The radon and/or thoron gas concentration of the source remains constant during the 'sampling' period in the direct diffusion process, and negligible in the outside region of the sensitive volume during the reverse diffusion process; and
2. Radon daughters and thoron daughters are essentially removed, i.e., filtered out by the membrane. This is true of any membrane, except for very high porosity media of little interest in the context of this paper.
3. Back-diffusion of radioactive gas is negligible during both the direct and

reverse diffusion processes.

With the geometry sketched in Fig. 1 with a source of radon or thoron of concentration N_1 in volume V_1, i.e., outside region, separated from the sensitive volume, V_2, by a membrane of thickness δ, surface area A and permeability k, one may write the kinetic equations describing the diffusion process as follows:

$$\frac{dN_1}{dt} = - \lambda_n N_1 + a - \frac{QN_1}{V_1} - \frac{kA}{\delta V_1} (N_1 - N_2) \qquad \text{Eq 1}$$

and

$$\frac{dN_2}{dt} = - \lambda_n N_2 + \frac{kA}{\delta V_2} (N_1 - N_2) \qquad \text{Eq 2}$$

where a is the production rate of the source (radon or thoron). The symbol λ_n stands for the radioactive decay constant of the gas (radon or thoron) and Q is the air flow in the outside region. It should be noted that the first, third and fourth terms on the right hand side of Eq. 1 describe, respectively, the removal of the radioactive gas in volume V_1 by radioactive decay, by forced ventilation or air flow, and by diffusion through the membrane. The first and second terms on the right hand side of Eq. 2 describe, respectively, the removal of radioactive gas in V_2 by radioactive decay and the increase in gas concentration in the same volume by diffusion through the membrane from V_1.

Equations 1 and 2 can be rearranged as follows:

$$\frac{dN_1}{dt} + MN_1 = a + SN_2 \qquad \text{Eq 3}$$

and

$$\frac{dN_2}{dt} + PN_2 = TN_1 \qquad \text{Eq 4}$$

where,

$$M = \Lambda + \frac{kA}{\delta V_1} , \quad \Lambda = \lambda_n + Q/V_1$$

$$S = \frac{kA}{\delta V_1} , \quad P = \lambda_n + \frac{kA}{\delta V_2} \quad \text{and} \quad T = \frac{kA}{\delta V_2}$$

1. DIRECT DIFFUSION PROCESS

The solution to the above equation can be simplifed in our case as for the outside region, $dN_1/dt = 0$, i.e., steady-state condition. After some algebraic manipulation, and taking into account the boundary conditions $N_1(t=0) = N_1$ and $N_2(t=0) = 0$, it is easy to show that:

$$N_2 = \frac{E}{K}\,(1-e^{-Kt}) \qquad \text{Eq 5}$$

where,

$$E = aT/M \quad \text{and} \quad K = P - (ST/M)$$

N_2 is Equation 5 reduces to a constant value, E/K, for large values of t. It can be shown:

$$\frac{E}{K} = \left(\frac{a}{\Lambda}\right) \frac{1}{1 + \left(\frac{\lambda_n \delta}{k}\right)\left(\frac{V_2}{A}\right)} \qquad \text{Eq 6}$$

The diffusion of radioactive gas into the sensitive volume is described by Equation 5. However, as soon as the gas enters the sensitive volume it decays into its decay products, i.e., daughters, for short. The differential equation that describes the growth of the daughter products can be written as:

$$\frac{dN_i}{dt} = N_{i-1}\,\lambda_{i-1} - N_i\lambda_i \qquad \text{Eq 7}$$

where the index i in Equation 7 refers to RaA, RaB, RaC for radon daughters and ThA, ThB, ThC and ThC' for thoron daughters. The index i-1 refers to the parent product of daughter product i, i.e., Rn-222 for RaA, and so on. N_i in Equation 7 can be solved according to standard methods of differential calculus. Notice that for i ≡ RaA or ThA, $N_{i-1} = N_2$ which is given by Equation 5. It should also be noted that the solution for N_i according to Equation 7, is then used to solve the differential equation describing the growth of its daughter product, and so on. Hence, the calculation increases considerably in complexity for subsequent daughter products.

It can be shown, after some algebraic manipulation, that the daughter concentrations are given by the following equations, where the indices A, B and C are used to denote, respectively, RaA or ThA, RaB or ThB and RaC or ThC. The symbol C' will be used to denote ThC'.

$$N_A = \lambda_n\left(\frac{E}{K}\right)\left[\frac{1}{\lambda_A} - \left(\frac{1}{\lambda_A} - \frac{1}{\lambda_A-K}\right)e^{-\lambda_A t} - \frac{1}{\lambda_A-K}\, e^{-Kt}\right] \qquad \text{Eq 8}$$

$$N_B = \lambda_n\lambda_A\left(\frac{E}{K}\right)\left[\frac{1}{\lambda_A\lambda_B} + X_2 e^{-\lambda_A t} + U_2 e^{-\lambda_B t} - Y_2\, e^{-Kt}\right] \qquad \text{Eq 9}$$

$$N_C = \lambda_n\lambda_A\lambda_B\left(\frac{E}{K}\right)\left[\frac{1}{\lambda_A\lambda_B\lambda_C} + X_1 e^{-\lambda_A t} + Y_1 e^{-\lambda_B t} - Z_1 e^{-Kt} + U_1 e^{-\lambda_C t}\right] \qquad \text{Eq 10}$$

$$N_{C'} = 0.663\, N_C(\lambda_C/\lambda_{C'}) \qquad \text{Eq 11}$$

where

$$X_2 = K/\lambda_A(\lambda_A-K)\ (\lambda_B-\lambda_A)$$

$$Y_2 = 1/(\lambda_A-K)\ (\lambda_B-K)$$

$$U_2 = Y_2 - X_2 - 1/\lambda_A\lambda_B$$

$$X_1 = K/\lambda_A(\lambda_A-K)\ (\lambda_B-\lambda_A)\ (\lambda_C-\lambda_A)$$

$$Y_1 = (1/\lambda_C-\lambda_B)\left[\frac{1}{(\lambda_A-K)\ (\lambda_B-K)} - \frac{1}{\lambda_A\lambda_B} - \frac{K}{\lambda_A(\lambda_A-K)\ (\lambda_B-\lambda_A)}\right]$$

$$Z_1 = 1/(\lambda_A-K)\ (\lambda_B-K)\ (\lambda_C-K)$$

$$U_1 = Z_1 - Y_1 - X_1 - 1/\lambda_A\lambda_B\lambda_C$$

The activity, A, corresponding to each decay product is simply given by the relation:

$$A_i = N_i\lambda_i \qquad \text{Eq 12}$$

Equations 8 to 11, and 12 give, respectively, the daughter concentration and activity as a function of time. However, of interest from a practical standpoint is the integrated activity during a given, predetermined, sampling time. (The same applies to the radioactive gas.) This is important for personal dosimetry and environmental monitoring applications as will be

discussed below. In this case a suitable detector and memory unit are used to detect and store radiation data, e.g., counts from the decay of the radioactive gas and its decay products. The integrated activity for the radioactive gas and its daughter products is given, respectively, by Equations 13 and 14:

$$IAS(n) = \int_{T_1}^{T_2} N_n \lambda_n dt \qquad \text{Eq 13}$$

$$IAS(i) = \int_{T_1}^{T_2} N_i \lambda_i dt \qquad \text{Eq 14}$$

where N_n is given by Equation 5. N_i is given by Equations 8 to 11. T_2 and T_1 are the limits of integration. Hence T_2-T_1 is the sampling time. Usually T_1 is taken as the beginning of sampling, i.e., $T_1=0$. The symbols i and n refer, respectively, to the gas and its daughter products. The symbol IAS stands for integrated activity during sampling.

Using Equation 5 and Equations 8 to 11, IAS(n) and IAS(i) can easily be calculated. If an α-particle detector (i.e., silicon-barrier or diffused-junction) is used to measure accumulated α-activity in the sensitive volume V_2, β-emitters need not be considered, e.g., RaB and ThB. It can be shown that:

$$IAS(n) = \lambda_n\left(\frac{E}{K}\right)\left[(T_2-T_1) + \frac{1}{K}(e^{-KT_2} - e^{-KT_1})\right] \qquad \text{Eq 15}$$

$$IAS(A) = \lambda_n\left(\frac{E}{K}\right)\left[(T_2-T_1) - \frac{K}{\lambda_A(\lambda_A-K)}(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) + \frac{\lambda_A}{K(\lambda_A-K)}(e^{-KT_2} - e^{-KT_1})\right] \qquad \text{Eq 16}$$

$$IAS(C) = \lambda_n\lambda_A\lambda_B\lambda_C\left(\frac{E}{K}\right)\left[\frac{T_2-T_1}{\lambda_A\lambda_B\lambda_C} - \frac{X_1}{\lambda_A}(e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) - \frac{Y_1}{\lambda_B}(e^{-\lambda_B T_2} - e^{-\lambda_B T_1}) + \frac{Z_1}{K}(e^{-KT_2} - e^{-KT_1}) - \frac{U_1}{\lambda_C}(e^{-\lambda_C T_2} - e^{-\lambda_C T_1})\right] \qquad \text{Eq 17}$$

$$IAS(C') = 0.663\ IAS\ (C) \qquad \text{Eq 18}$$

2. REVERSE DIFFUSION PROCESS

If at a given time T_s the sensitive volume V_2 is removed from the outside region V_1 to an area of negligible radon and thoron gas concentration, it can be shown from Equation 4 with $N_1=0$ at $t \geq T_s$ that the expression describing the concentration of radioactive gas in V_2 is:

$$N_2 = \frac{E}{K}(1-e^{-KT_s})\ e^{-Pt} = N_n(T_s)\ e^{-Pt} \qquad \text{Eq 19}$$

where $N_n(T_s) = E/K\ (1 - e^{-KT_s})$ is a constant for a given T_s.

Following a procedure similar to that for the direct diffusion process, it can be shown:

$$N_A = \lambda_n N_n(T_s) \left(\frac{1}{\lambda_A - P}\right) (e^{-Pt} - e^{-\lambda_A t}) + N_A(T_s)\ e^{-\lambda_A t} \qquad \text{Eq 20}$$

where $N_A(T_s)$ is N_A for the direct diffusion case for a sampling time T_s, i.e., Equation 8 for $t=T_s$.

$$N_B = \lambda_n \lambda_A N_n(T_s) \left(\frac{1}{\lambda_A - P}\right) \left[\frac{e^{-Pt}}{\lambda_B - P} - \frac{e^{-\lambda_A t}}{\lambda_B - \lambda_A} - \frac{(P-\lambda_A)\ e^{-\lambda_B t}}{(\lambda_B - P)\ (\lambda_B - \lambda_A)}\right]$$

$$+ N_A(T_s)\ \frac{\lambda_A\ e^{-\lambda_A t}}{\lambda_B - \lambda_A} + \left(N_B(T_s) - \frac{N_A(T_s)\lambda_A}{\lambda_B - \lambda_A}\right)\ e^{-\lambda_B t} \qquad \text{Eq 21}$$

where $N_B(T_s)$ is N_B for the direct diffusion case for a sampling time T_s, i.e., Equation 9 for $t=T_s$.

$$N_C = \lambda_n \lambda_A \lambda_B N_n(T_s) \left(\frac{1}{\lambda_A - P}\right) \left[A(2)\ e^{-Pt} - B(2)\ e^{-\lambda_A t} - C(2)\ e^{-\lambda_B t}\right]$$

$$+ D(2)\ e^{-\lambda_A t} + E(2)\ e^{-\lambda_B t}$$

$$+ \left[N_C(T_s) - D(2) - E(2) - \lambda_n \lambda_A \lambda_B N_n(T_s) \left(\frac{1}{\lambda_A - P}\right)\left(A(2)-B(2)-C(2)\right)\right] e^{-\lambda_C t} \qquad \text{Eq 22}$$

where $N_C(T_s)$ is N_C for the direct diffusion case for a sampling time T_s, i.e., Equation 10 for $t=T_s$.

$$N_C' = 0.663\ N_C(\lambda_C/\lambda_C') \qquad \text{Eq 23}$$

where

$$A(2) = 1/(\lambda_C - P)\ (\lambda_B - P)$$

$$B(2) = 1/(\lambda_C - \lambda_A)\ (\lambda_B - \lambda_A)$$

$$C(2) = (P - \lambda_A)/(\lambda_C - \lambda_B)\ (\lambda_B - P)\ (\lambda_B - \lambda_A)$$

$$D(2) = N_A(T_s)\lambda_A\lambda_B/(\lambda_C - \lambda_A)\ (\lambda_B - \lambda_A)$$

$$E(2) = (\lambda_B/\lambda_C - \lambda_B)\left[N_B(T_s) - \frac{N_A(T_s)\lambda_A}{\lambda_B - \lambda_A}\right]$$

As for the case of the direct diffusion process, the activity is given by Equation 12.

Similarly, the integrated activity for the reverse diffusion case can be derived following a similar procedure as that for the direct case. Hence,

$$IAS(n) = N_n(T_s)\ (\lambda_n/P)\ (e^{-PT_1} - e^{-PT_2}) \qquad \text{Eq 24}$$

$$IAS(A) = \frac{\lambda n \lambda A N n(T_s)}{\lambda_A - P}\left[-\frac{1}{P}(e^{-PT_2} - e^{-PT_1}) + \frac{1}{\lambda_A}\ (e^{-\lambda_A T_2} - e^{-\lambda_A T_1})\right]$$

$$- N_A(T_s)\ (e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) \qquad \text{Eq 25}$$

$$IAS(C) = \lambda_n\lambda_A\lambda_B\lambda_C N_n(T_s)\left(\frac{1}{\lambda_A - P}\right)\left[-\frac{A(2)}{P}\ (e^{-PT_2} - e^{-PT_1})\right.$$

$$\left. + \frac{B(2)}{\lambda_A}\ (e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) + \frac{C(2)}{\lambda_B}\ (e^{-\lambda_B T_2} - e^{-\lambda_B T_1})\right]$$

$$- \frac{\lambda_C D(2)}{\lambda_A}\ (e^{-\lambda_A T_2} - e^{-\lambda_A T_1}) - \frac{\lambda_C E(2)}{\lambda_B}\ (e^{-\lambda_B T_2} - e^{-\lambda_B T_1})$$

$$- \left[N_C(T_s) - D(2) - E(2) - \lambda_n\lambda_A\lambda_B N_n(T_s)\left(\frac{1}{\lambda_A - P}\right)\right.$$

$$\left.\left(A(2) - B(2) - C(2)\right)\right]\left(e^{-\lambda_C T_2} - e^{-\lambda_C T_1}\right) \qquad \text{Eq 26}$$

$$IAS(C') = 0.663\ IAS(C) \qquad \text{Eq 27}$$

It should be noted that T_1 and T_2 in Equations 24 to 26 refer now to the time after the end of the 'sampling' period, i.e., end of direct diffusion process. Hence, $T_1 > T_s$ and $T_2 > T_1$ where T_s is taken as 0 relative to the time elapsed during the reverse diffusion process.

If for practical reasons a silicon barrier or diffused-junction α-detector is used in the sensitive volume, it can be shown that the total α-count, $N_{\alpha,T}$, recorded by the detector and associated electronic circuitry is given by:

$$N_{\alpha,T} = 2.22\ \varepsilon\eta\delta\theta\left[\left\{IAS(n) + \Sigma_i\ IAS(i)\right\}_{dp} + \left\{IAS(n) + \Sigma_i\ IAS(i)\right\}_{rp}\right]$$

$$= 2.22\ \varepsilon\eta\delta\theta\sum_{dp,rp}\left[IAS(n) + \Sigma_i\ IAS(i)\right] \qquad \text{Eq 28}$$

where the first two terms and the last two terms on the right hand side of Equation 28 refer to the direct diffusion process and reverse diffusion process, respectively. The indices dp and rp stand, respectively, for direct and reverse diffusion process.

In Equation 28, 2.22 is a numerical factor to convert pCi into disintegrations per minute (dpm); ε is the α-counting efficiency of the detector; $\eta(<1)$ is a factor which takes into account absorption of α-particles in Mylar and other materials normally used to protect the detector's surface; $\delta(<1)$ takes into consideration plate-out of daughter products on the inner surface of the membrane; and θ is a factor which takes into account electrostatic effects, e.g., α-collection enhancement by electrostatic means, as used in some instrumentation (7).

If the characteristics of the membrane are known, the radioactive gas and daughter product concentrations can be estimated using Equation 28. Conversely, if the radioisotope concentrations are known, the characteristics of the membrane can be obtained.

THEORETICAL BACKGROUND (TIME-DEPENDENT CASE)

The expressions given above have been derived under the assumption of steady-state radiation conditions, i.e., the ^{222}Rn and ^{220}Rn concentration in V_1 remain constant during the sampling period (direct diffusion case) and during the reverse diffusion case, i.e., $N_1=0$ in Equation 4 for $t>\bar{T}_s$. However, radiation conditions in the field are usually time-dependent. It is, therefore, important to investigate the solutions to the above equations under well-defined, non steady-state, i.e., time-dependent, radiation conditions. Cases of practical interest are those in which radiation levels vary in a reasonably predictable fashion characteristic of field conditions, i.e., sinusoidally, linearly, exponentially and step-wise.

Variation in radiation levels can be brought about by variations in the source term, i.e., production rate, a, and/or the ventilation rate, i.e., air flow conditions, described by Q (see Equation 1). However, sometimes it is sufficient to indicate that N_1 in Equations 1 and 2 is time-dependent. This dependency on time is a direct reflection of the time-dependency characteristics of a and/or Q. Calling X any one of the above variables, one may write, in general, the following expression:

$$X(t) = X_0 F(t) \qquad \text{Eq 29}$$

where, the function F(t) can assume any time-dependent analytical form, and can be thought of as a perturbation introduced in the otherwise undisturbed, steady-state, condition, i.e., X_0. Cases of interest in the context of this paper are:

$$F(t) = 1 + a' \sin \omega t \quad \text{(sinusoidal case)} \qquad \text{Eq 30}$$

$$F(t) = 1 + \eta t \quad \text{(linear case)} \qquad \text{Eq 31}$$

$$F(t) = e^{\delta t} \quad \text{(exponential case)} \qquad \text{Eq 32}$$

$$\left.\begin{aligned} F(t) &= F_0 \text{ for } t \leq t_0, \text{ and} \\ F(t) &= q\,F_0 \text{ for } t > t \\ \text{where,} \quad & q \neq 1, \text{ but } q \gtrless 0 \end{aligned}\right\} \qquad \text{Eq 33}$$

The symbols a', η, δ and q are the perturbation amplitudes corresponding to Equations 30 to 33, respectively. For a' = 0, η = 0, and δ = 0, F(t) = 1, i.e., non-perturbed, or steady-state case.

Analytical solutions under time-dependent conditions can be obtained by substituting Equation 29, taking into account Equations 30 to 33, into Equations 1 and 2. This has been done in part II of this paper (8), and this subject will not be dealt with here any further.

FINAL REMARKS

The analytical expressions derived in this paper find practical application in personal dosimetry and in environmental monitoring. A thorough analysis and graphical representation of the above equations, and of the equation derived for the time-dependent radiation case, i.e., Part II, have been presented elsewhere (9).

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